

Quantum theory of photonic crystal polaritons

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We formulate a full quantum mechanical theory of the interaction between electromagnetic modes in photonic crystal slabs and quantum well excitons embedded in the photonic structure. We apply the formalism to a high index dielectric layer with a periodic patterning suspended in air. The strong coupling between electromagnetic modes lying above the cladding light line and exciton center of mass eigenfunctions manifests itself with the typical anticrossing behavior. The resulting band dispersion corresponds to the quasi-particles coming from the mixing of electromagnetic and material excitations, which we call *photonic crystal polaritons*. We compare the results obtained by using the quantum theory to variable angle reflectance spectra coming from a scattering matrix approach, and we find very good quantitative agreement.

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Introduction. Since the pioneering works of more than fifteen years ago by E. Yablonovitch and S. John [1], a great deal of research in physics has been dedicated to the study of the optical properties of photonic crystals [2]. In the last few years much attention has been devoted to photonic crystal (PC) structures embedded in planar dielectric waveguides, which are also called *photonic crystal slabs*. These are systems with a periodic dielectric response in the plane of the waveguide, while a dielectric mismatch is used to confine the electromagnetic field in the vertical direction.

One of the main issues of PC slabs is the existence of the light line problem: only photonic modes lying below the cladding light line are truly guided and with zero intrinsic linewidth, while the modes lying above the light line are radiative. A method to calculate the photonic band dispersion and the intrinsic linewidth (complex energies) both below and above the light line has been recently proposed [3, 4]. Until now, however, the physics of PCs has been considered mostly for what concerns the “optical” point of view, that is studying the propagation of light in periodic dielectric media.

In this work we analyze the radiation-matter interaction in PC slabs, by considering the effects of the interplay between the electromagnetic field and semiconductor quantum well (QW) excitons. In particular, we show that polaritonic effects are present in PC slabs when the exciton-photon coupling is larger than the intrinsic radiative linewidth of a photonic mode above the cladding light line. The band dispersion is greatly modified in the vicinity of the excitonic resonance, leading to the formation of mixed states which are analogous to exciton-polaritons in bulk semiconductors [5] and microcavities [6–8] and which we call *photonic crystal polaritons* (PCPs). Experimental results showing the formation of PCPs in organic-based systems were previously reported in Ref. 9.

Theory. Here we briefly describe the quantum theory of PCPs, starting with the second quantized total

hamiltonian of the system, which is given by

$$\begin{aligned} \hat{H} = & \sum_{\mathbf{k},n} \hbar\omega_{\mathbf{k}n} \hat{a}_{\mathbf{k}n}^\dagger \hat{a}_{\mathbf{k}n} + \sum_{\mathbf{k},\nu} \hbar\Omega_{\mathbf{k}\nu} \hat{b}_{\mathbf{k}\nu}^\dagger \hat{b}_{\mathbf{k}\nu} \\ & + i \sum_{\mathbf{k},n,\nu} C_{\mathbf{k}n\nu} (\hat{a}_{\mathbf{k}n} + \hat{a}_{-\mathbf{k}n}^\dagger) (\hat{b}_{\mathbf{k}\nu}^\dagger - \hat{b}_{-\mathbf{k}\nu}) \\ & + \sum_{\mathbf{k},\nu,n_1,n_2} \frac{C_{\mathbf{k}n_1\nu}^* C_{\mathbf{k}n_2\nu}}{\hbar\Omega_{\mathbf{k}\nu}} (\hat{a}_{-\mathbf{k}n_1} + \hat{a}_{\mathbf{k}n_1}^\dagger) (\hat{a}_{\mathbf{k}n_2} + \hat{a}_{-\mathbf{k}n_2}^\dagger) \end{aligned} \quad (1)$$

In Eq. (1), the first term indicates the photonic band dispersion (real part of the complex eigenenergies), $\hat{a}_{\mathbf{k}n}$ ($\hat{a}_{\mathbf{k}n}^\dagger$) being the destruction (creation) operators of a photon with wave vector \mathbf{k} and band number n . These energies are obtained by expanding the magnetic field in terms of the guided modes of the effective waveguide, that is the one with an average dielectric constant, and then by solving the Maxwell equation as a linear eigenvalue problem [3]. The imaginary part of the photonic modes is calculated by using a perturbative approach [4]. If we restrict our considerations to the system schematically shown in the inset of Fig. 1a, the direction of periodicity is x and thus the wave vector is given by the component k_x . The modes in a PC slab suspended in air can be classified as even (odd) with respect to specular reflection through the plane xy , and even (odd) with respect to the plane of incidence (that is xz in the case considered here). Throughout this paper we consider only modes which are spatially even with respect to xy plane, and odd with respect to the vertical mirror plane (even TE modes). These modes interact with transverse QW excitons. The excitonic problem, whose solution gives the energies $\hbar\Omega_{\mathbf{k}\nu}$, is treated by solving the Schrödinger equation for the exciton center of mass envelope function in a periodic piecewise constant potential having the same patterning as the photonic crystal structure. The in-plane wave vector and an integer ν are good quantum numbers for the exciton wavefunctions, due to the periodic potential and to the spatial dispersion of the center

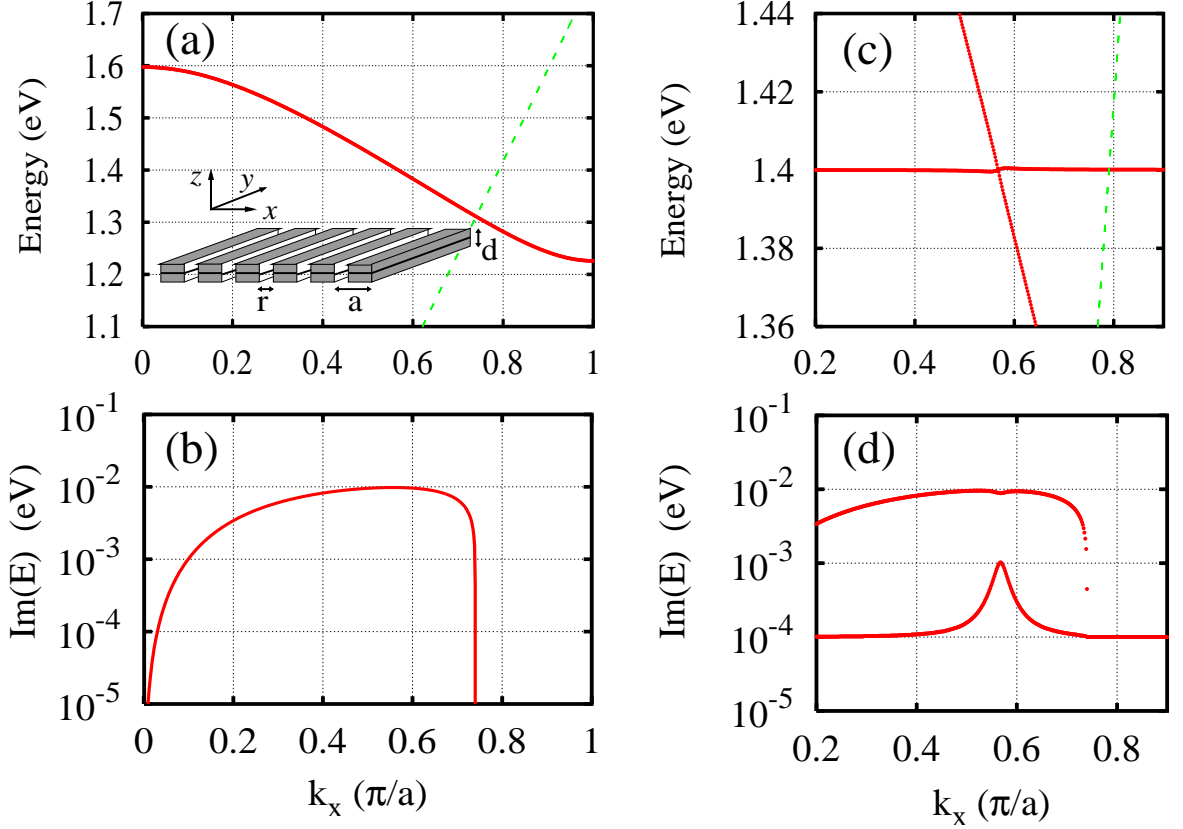


FIG. 1: (a) Real and (b) imaginary parts of the complex photonic energies of the even TE mode as a function of the wave vector in the first Brillouin zone, for the system shown in the inset, without excitonic resonance (no QW); (c) real and (d) imaginary parts for the same structure with a QW at the center of the dielectric core, with an excitonic resonance at $\hbar\Omega_0 = 1.4$ eV. The dashed lines in (a) and (c) represent the light line in air.

of mass; the corresponding destruction (creation) operators are $\hat{b}_{\mathbf{k}\nu}$ ($\hat{b}_{\mathbf{k}\nu}^\dagger$). The exciton-photon coupling matrix elements, $C_{\mathbf{k}n\nu}$, are calculated in terms of the microscopic physical quantities as

$$C_{\mathbf{k}n\nu} = \left(\frac{2\pi e^2 \hbar \Omega_{\mathbf{k}\nu}^2}{\omega_{\mathbf{k}n}} \right)^{1/2} \langle \Psi_{\mathbf{k}\nu}^{(exc)} | \sum_j \mathbf{E}_{\mathbf{k}n}(\mathbf{r}_j) \cdot \mathbf{r}_j | 0 \rangle. \quad (2)$$

In Eq. (2), $\Psi_{\mathbf{k}\nu}^{(exc)}$ is the all-electron exciton wavefunction, $\mathbf{E}_{\mathbf{k}n}$ is the electric field profile for the photonic mode at frequency $\omega_{\mathbf{k}n}$ in the PC slab, and the sum is over all the QW electrons. In particular, $C_{\mathbf{k}n\nu}$ is found to depend on the overlap between the exciton envelope function and the transverse electric field (for what concerns TE modes). It is proportional to $(f/S)^{1/2}$, where f/S is the oscillator strength per unit area which depends on the QW thickness [8]. The hamiltonian is diagonalized by using a generalized Hopfield transformation to expand new destruction (creation) operators $\hat{P}_{\mathbf{k}}$ ($\hat{P}_{\mathbf{k}}^\dagger$) as a linear combination of $\hat{a}_{\mathbf{k}n}$ ($\hat{a}_{\mathbf{k}n}^\dagger$) and $\hat{b}_{\mathbf{k}\nu}$ ($\hat{b}_{\mathbf{k}\nu}^\dagger$), with the condition $[\hat{P}_{\mathbf{k}}, \hat{H}] = E_{\mathbf{k}} \hat{P}_{\mathbf{k}}$ [10]. The new eigenenergies $E_{\mathbf{k}}$

correspond to mixed excitations of radiation and matter, i. e. the PCPs.

All the results presented in this paper refer to a model structure, namely a high index dielectric core suspended in air with a one dimensional patterning, and a QW placed at the center of it (see inset of Fig. 1a). The dielectric constant of the semiconductor-based core layer is set to the value $\varepsilon = 12$, the dielectric constant of the air being simply $\varepsilon_{air} = 1$. The slab thickness and the air fraction are set to the values $d/a = 0.2$ and $r/a = 0.3$ respectively, the lattice constant being $a = 350$ nm. A QW of thickness $L_{QW} = 8$ nm is considered, and the oscillator strength per unit area is set to the typical value $f/S = 8.4 \times 10^{12} \text{ cm}^{-2}$. The intrinsic radiative exciton linewidth is assumed to be $\Gamma = 0.1$ meV.

Results and Discussion. In Fig. 1a we show the photonic band dispersion in the energy range 1.1-1.7 eV. In Fig. 1b we display the corresponding imaginary part as a function of the wave vector, which shows a maximum at about 1.4 eV. The imaginary part goes to zero at $k_x = 0$, that is at normal incidence, and at $k_x = 0.74$; this last behavior is due to the crossing of the light line,

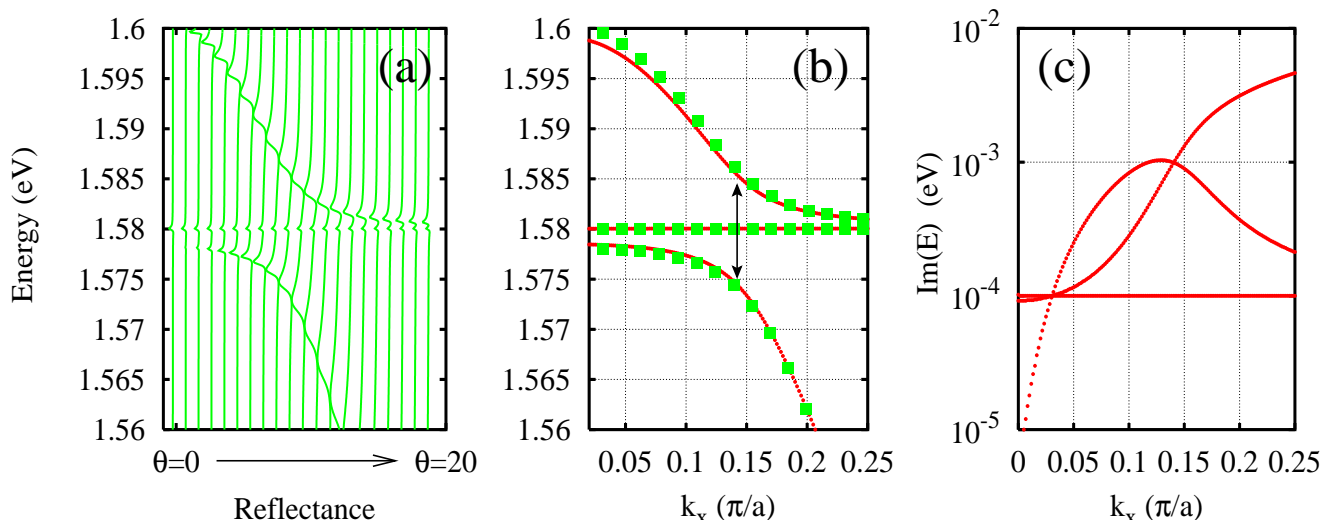


FIG. 2: (a) Calculated surface reflectance of a TE polarized plane wave incident on the structure of the inset of Fig. 1a. The excitonic resonance is set at $\hbar\Omega_0 = 1.58$ eV; the angle of incidence is varied from $\theta = 0^\circ$ to $\theta = 20^\circ$, with a step of 1° . (b) Real and (c) imaginary parts of the complex energies of the photonic crystal polaritons, calculated with the quantum theory. The squared points in (b) are extracted from the calculated reflectance curves shown in (a), and the polariton splitting is indicated by an arrow.

as shown in Fig. 1a, corresponding to the photonic mode becoming truly guided and stationary. If the photonic imaginary part is larger than the exciton-photon coupling matrix element (which is of the order of a few meV), a QW placed at the center of the PC slab does not produce any important change in the photonic band dispersion. Indeed, this result is shown in Figs. 1c and 1d, in which we display the real and imaginary parts of the complex eigenenergies coming from the diagonalization of Eq. (1) with an excitonic resonance at $\hbar\Omega_0 = 1.4$ eV. In this *weak coupling* regime the photon and the exciton are almost uncoupled, as it can be seen from the crossing of the two dispersion relations in Fig. 1c. The imaginary part of the exciton increases by an order of magnitude correspondingly to the crossing point, but this has a negligible effect on the photonic radiative linewidth, which is still an order of magnitude larger than the excitonic one (see Fig. 1d).

In order to observe the *strong coupling* regime, the energy of the excitonic resonance has to lie where the imaginary part of the photonic mode is smaller than the coupling matrix element. From Fig. 1a and 1b we see that for $\hbar\Omega_0 = 1.58$ eV the imaginary part of the corresponding photonic band is about $\text{Im}(\hbar\omega) = 10^{-3}$ eV. In Fig. 2a we show the results of a variable angle reflectance calculation, which is done by a scattering matrix approach [11]. The dielectric function of the QW layer is frequency dependent, with a resonance at $\hbar\Omega_0 = 1.58$ eV. It is known that the sharp features appearing in the reflectance spectrum correspond to the excitation of photonic modes above the light line [9, 12], thus giving a

point (\mathbf{k}, ω) of the corresponding photonic band dispersion; the wave vector component parallel to the surface is given by $k = (\omega/c) \sin \theta$. The (\mathbf{k}, ω) points extracted from the results of Fig. 2a are compared to the dispersion of PCPs in Fig. 2b, in which the squares are the scattering matrix results. The quantum theory is in excellent agreement with the classical approach. In particular, both theories confirm the anticrossing behavior of exciton and photon modes, which is a clear effect of the strong coupling regime. With the parameters used in this work, the polariton splitting is as high as 10 meV at the anticrossing point $k_x = 0.14$, as shown in Fig. 2b. This splitting is found to be slightly larger than in semiconductor-based microcavities [7, 8]. In Fig. 2c, finally, the imaginary part of the PCPs complex eigenenergies is shown. At $k_x = 0.14$ the imaginary parts of the upper and lower polariton branches become equal to the same value $\text{Im}(E) = 10^{-3}$ eV, thereby indicating that mixed states of radiation and matter form in the PC slab. The dispersionless curve at $\text{Im}(E) = 10^{-4}$ eV in Fig. 2c corresponds to the uncoupled excitonic modes.

Conclusions. We have shown that the quantum theory of the interaction between electromagnetic modes in photonic crystal slabs and QW excitons can describe both the weak and strong coupling regimes. Moreover, we have found a very good agreement between the quantum theory and a classical approach based on a scattering matrix method. We thus conclude that new mixed states of radiation and matter excitations can be experimentally measured in semiconductor-based photonic crystal slabs by using variable angle reflectance or transmittance

techniques, provided that the experimental conditions required for being in the strong coupling regime could be satisfied.

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